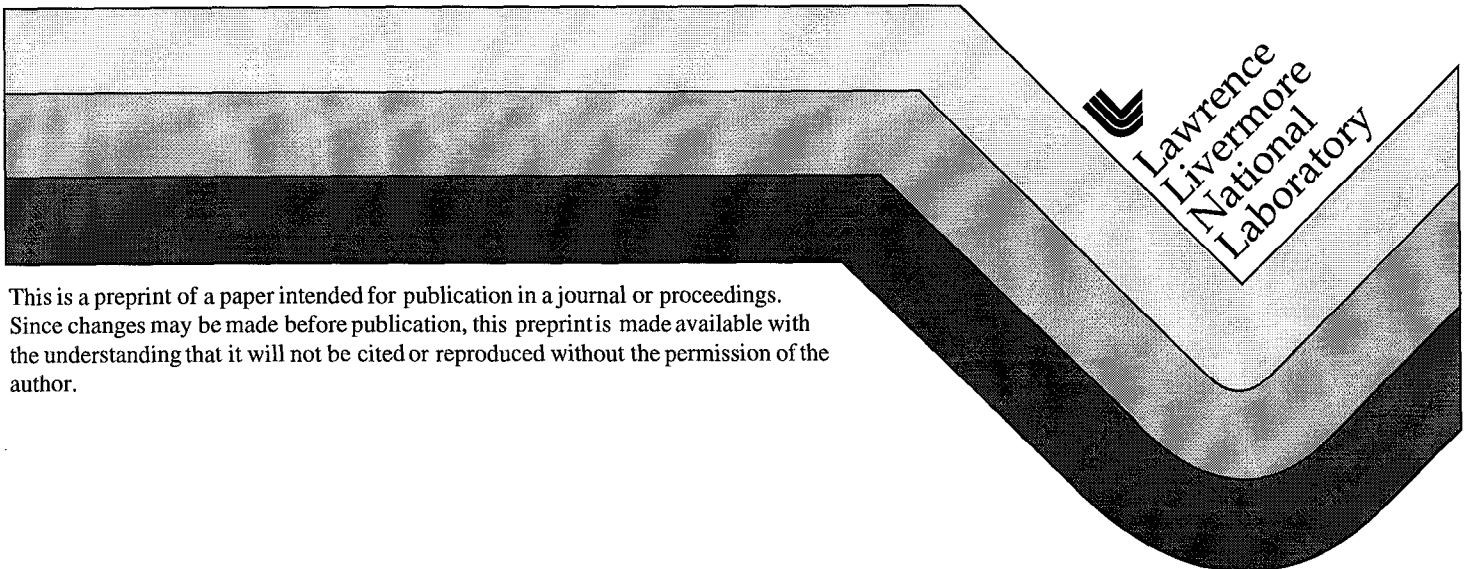# Cryogenic D-T Fuel Layers Formed in 1 MM Spheres by Beta-Layering

J. Sater
B. Kozioziemski
G. W. Collins
E. R. Mapoles
J. Pipes
J. Burmann
T. P. Bernat

October 15, 1998

CRYOGENIC D-T FUEL LAYERS FORMED IN 1 MM SPHERES BY BETA-LAYERING

J. Sater[†], B. Kozioziemski[‡], G.W. Collins[‡], E.R. Mapoles[‡], J. Pipes[*], J. Burmann[†], T.P. Bernat[‡]

[†]Schafer Corporation
303 Lindbergh Ave., Livermore, Ca. 94550

[*]Allied Signal Inc.
2021 Las Positas Ct., Livermore, Ca. 94550

[‡]Lawrence Livermore National Laboratory
P.O. Box 808, Livermore, CA 94550

ABSTRACT

Solid D-T fuel smoothly layered on the interior of spherical capsules is required for all inertial confinement fusion ignition target designs. One process for forming these layers, beta-layering, has been studied in surrogate geometries such as open cylinders or tori to allow accurate characterization of the DT surfaces. We present the first results from beta layering in 1 mm spherical containers, such as will be used in upcoming Omega experiments. These results are also directly relevant to ignition capsules for the National Ignition Facility. We find that layers can form with roughness as small as 1.2 microns rms, and that results are strongly dependent upon freezing rate as well as layer thickness.

INTRODUCTION

All high gain target designs for inertial confinement fusion (ICF) employ uniform layers of condensed DT to achieve efficient ignition of the fuel [1]. Various routes to the formation of these layers within target shells have been investigated [2], and the most promising method relies on the radioactive self heating of condensed DT to redistribute the solid along the isotherms in the fuel container [3], [4], [5]. This process is referred to as "beta layering". The resulting roughness of the ice surface has been characterized optically and been found to be in the range of 1 - 1.5 μm RMS for cylindrical layers approximately 100 μm thick and 2 mm in diameter. It is desirable to decrease this roughness to reduce the effects of mix during the implosion of an ICF capsule and increase the drive flexibility available to experimental designers [6].

When liquid DT freezes in an isothermal shell, it initially forms a very rough layer on the bottom of the container. Because the ice heats itself with a volumetric rate of $q_s = 5.06 \times 10^{-2}$ W/cm^3, the surface temperature increases with layer thickness. Thicker areas of the layer are warmer and consequently have higher vapor pressures. Thus, material sublimes away from thick regions and recondenses on thinner areas. If the container is isothermal, this leads to an ice surface that roughly conforms to the container surface. For pure DT, the rate at which the material migrates is $R_{DT} = q_s / s \rho_s$ [5], where s is the heat of sublimation (1580 J/mol), and ρ_s (5.03 x 10^{-2} mol/cm^3) is the density of the solid, so that $1/R_{DT} = 27$ minutes.

We present our recent results from beta layering in 1 mm spherical plastic containers. Our results indicate that layers can form with roughness as small as 1.2 microns rms. We also find that roughness is strongly dependent upon cooling rate and ice layer thickness.

EXPERIMENTAL DETAILS

Experiments are conducted on a mixture of approximately 25% T_2, 25% D_2, and 50% DT which is referred to as D-T. The sample is introduced as a liquid in the normal rotational state. J=1 to J=0 conversion occurs continuously throughout the experiment but no measurements are made of rotational content. Our experimental setup, data collection, and analysis is similar to that described in a recent paper [7].

The main difference is the replacement of a cylindrical sapphire sample cell with a series of 1mm diameter plastic

shells with 10 to 20 μm thick walls. The cell is filled with D-T liquid via a fill tube as pictured in Fig. 1. This design allows study of layering in a system very similar to what is required for many ICF target designs.
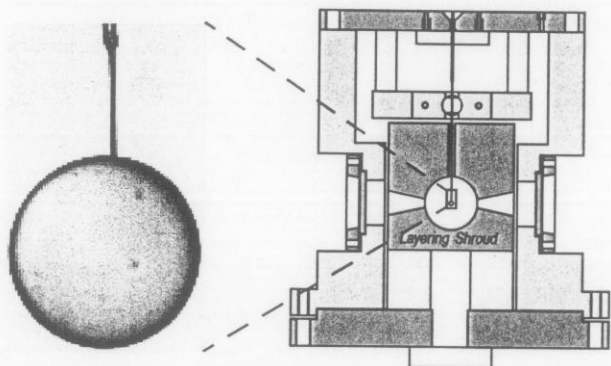

FIGURE 1. Layering occurs in a spherically symmetric geometry

A D-T ice surface conforms to the isotherms shaped by the surrounding layering shroud. Since a smooth spherical shape is desired, the layering shroud is a 25.4 mm diameter hollow sphere made of OFHC copper shown in Fig. 1. This provides a constant temperature spherical boundary condition. A few torr of ^{4}He gas inside the sphere causes conductive cooling of the plastic shell. The cavity is cooled from the bottom to minimize convection inside the sphere.

The plastic shell is placed at the center of the hollow sphere. A pair of 3 millimeter holes and a pair of 5 millimeter holes are bored in the layering shroud on perpendicular axes to provide a view of the sample cell. They have negligible effect on the spherical isotherms. The shroud is placed inside of a vacuum sealed secondary container.

Temperature is measured by use of calibrated germanium resistance thermometers(GRTs) placed on the top and bottom of the copper cavity. The mean value of the two GRTs is the reported temperature. Temperature stability is better than 2mK over a period of hours and temperature resolution is 1 mK. Calibration has been checked by measuring the triple point of our D_2 sample to be 18.72 K as compared to 18.73 K reported for nD_2 [8].

EXPERIMENTAL RESULTS

Fig. 2 shows the temperature and RMS surface roughness σ evolution during the course of an experiment carried out on a 3 day old sample of DT. σ is calculated from modes 2 through 128. In this experiment we cool slowly through the triple point at a constant rate of 0.5 mK/min until the set point temperature of 19.78K is reached. The temperature is then held constant for a period of time. The DT layer grows during the temperature ramp with a single growth front that begins near the top of the shell. The crystal growth front converges at the bottom of the shell to complete the layer. After formation, T = 19.78K, time is 295 minutes, and σ = 2.65 μm. The layer continues to smooth reaching a minimum σ of 1.15 μm at about 383 minutes. Afterwards the layer roughens. This roughening behavior is not well understood.
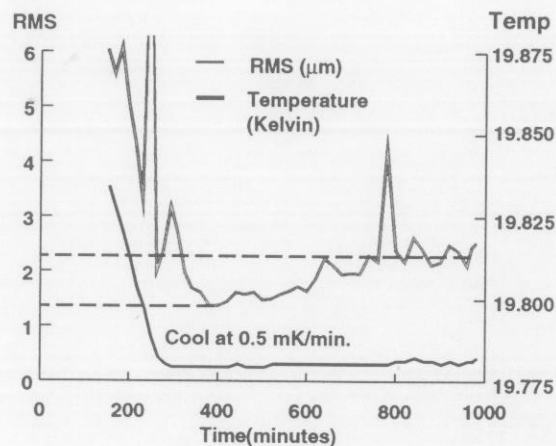

FIGURE 2. We observe that the layer smooths to a minimum value and then roughens with longer times.

Figure 3 shows the power spectral density for the layer at 400 minutes and at 1000 minutes. It is apparent that the layer at 400 minutes is smoother at all mode numbers

except perhaps at a singular point at mode 7. Most of the difference between the two layers occur in modes 2-4.
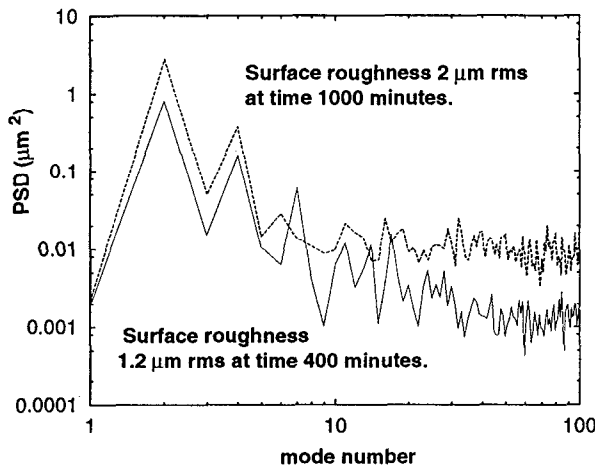

FIGURE 3. Increase in roughness over time after minima is largely due to effects at modal numbers between 2 and 4 though the largest relative increase occurs above mode 20.

The freezing rate has a strong effect on layer smoothness. σ is plotted in Fig. 4 for a series of 210 μm layers formed at different freezing rates. For rates slower than 3mK/min a dramatically smoother layer is produced. At faster freezing rates, the DT nucleates several small crystals instead of growing with a single front. Layers with several distinctly observable crystals are significantly rougher than those formed from a single nucleation site.

Data indicates that σ increases with decreasing layer thickness. In Fig. 5 we have collected all of our layering data with cooling rates between 1 and 2 mK/min and layer thicknesses between 150 and 225 μm. The 4.2 μm point was for a layer formed at the upper end of the cooling rate. Fig. 6 contains the power spectral density for a 152 μm layer and a 220 μm layer. The thicker layer is substantially smoother at all mode numbers. This could be due to the observed relative ease that thick layers nucleate as compared to thin layers. A contributing factor is the increase in variation of temperature near the ice/gas interface with thicker layers, i.e. $\delta T_{bump} = Q\, h\, \delta h_{bump}/k$. δT is the variation from the average temperature at the ice/gas interface. δh is the variation from the mean ice thickness.

The second type of distinct experiment carried out was a slow continuous freeze through the triple point of DT to a point well below the triple point. In Fig. 7 the temperature

vs. time evolution and σ vs. time are plotted on the same graph. Here we again observe that the surface reaches a minimum roughness and then at later times becomes quite rough as the layer continues to cool.
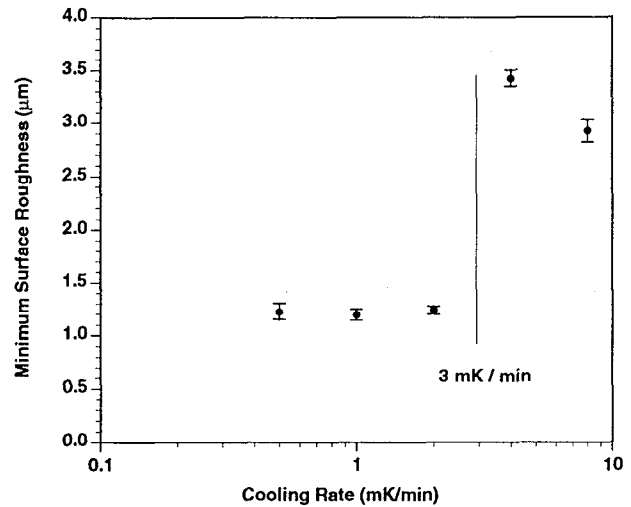

FIGURE 4. Plot of best surface roughness achieved for a series of 210 μm layers created with different cooling rates through the triple point of D-T.
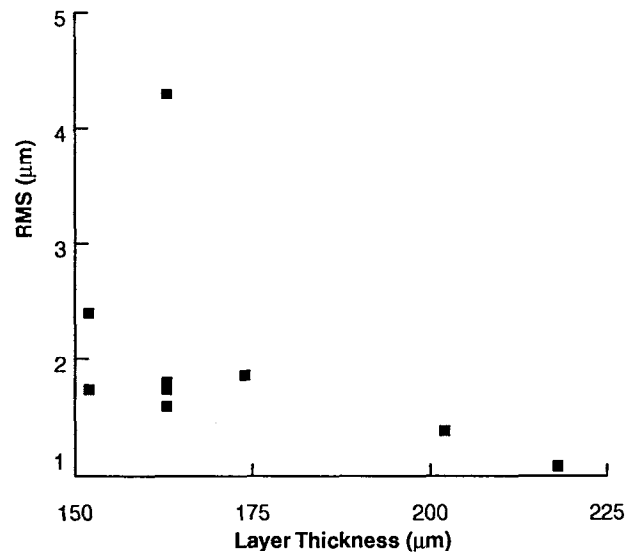

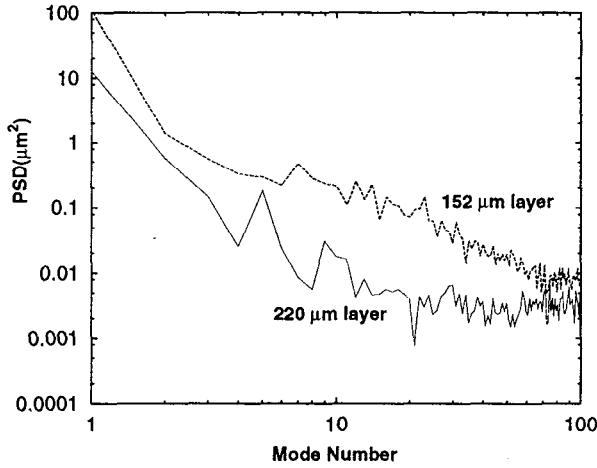**FIGURE 5. Thinner ice layers have a larger σ.**

FIGURE 6. The power spectral density plots indicate that a large amount of excess power for the thinner layers are in the middle part of the modal spectrum.

Layers at temperatures below T_{TP} - 0.5K are typically rougher than layers produced just below T_{TP}. Currently, NIF ignition target designs contain 80 μm thick D-T layers at 18K inside a 2 mm OD capsule. Our best DT layers are produced just below T_{TP} at about 19.7K. The gas density inside of the capsule is $\rho_{gas} = 0.7$mg/cc instead of the specified $\rho_{gas} = 0.3$ mg/cc, and thus would change the convergence of the ignition capsule design.

THEORY

The equilibrium ice layer minimizes the total free energy at constant volume. The thermal gradient resulting from the tritium decay heating favors uniform layers, however, the anisotropic surface energy favors facets. The curved shell geometry prevents the layer from having a single low energy facet exposed along the entire surface, and any recrystallization to reorient the surface is limited by grain boundary energy. We can model each of these energies to understand the layer roughness.

The total free energy per unit length is expressed as:

$$E_T = \int_A C\rho T dA + \sum_i^n \gamma^i{}_s L_i + \sum_i \gamma_{gb}{}^i t_i, \quad (1)$$

where C is the specific heat, ρ is the ice density, γ_s is the orientation dependent surface energy, L is the length of the crystal facet, γ_{gb} is the grain boundary energy, t is the grain boundary length, and A is the bulk ice area perpendicular to the cylindrical axis.
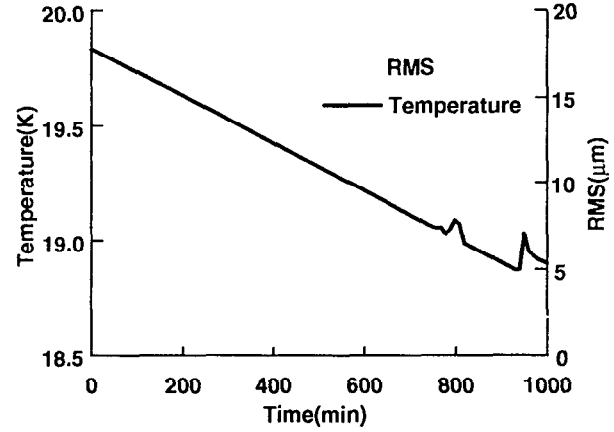

FIGURE 7. With a continuous freeze we also observe that the layer reaches a minimum σ early and then degrades.

The c - facet for solid deuterium was determined to have $\gamma = 6 \times 10^{-3}$ J/m^2 [9]. The best estimate for $\gamma_{gb} = 0.6 \times 10^{-3}$ J/m^2, 10 percent of the surface energy [10]. Using a one dimensional heat flow model for a layer with cylindrical geometry and n regular low energy facets, we find n = 53 minimizes the free energy for a 210 μm layer. The corresponding rms surface roughness is 0.15 microns. The model predicts the rms surface roughness decreases with increasing layer thickness out to 300 microns for a 1 mm shell, consistent with the data in Fig. 5.[a] The magnitude of the predicted roughness is a factor of 10 smaller than the data and is due to the highly idealized nature of the model. A more detailed model should include multiple crystal orientations and grain boundary energies.

---

[a]It should be noted that DT experiences a 12% shrinkage when cooling through the triple point. Thus a 1mm capsule full of liquid at just above the triple point would have a uniform solid layer 253 μm thick just below the triple point temperature.
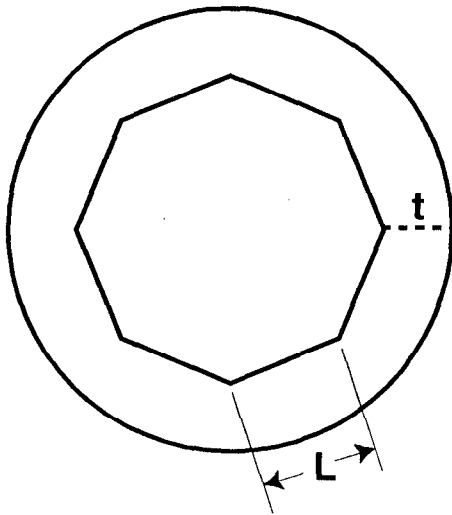
FIGURE 8. Simplified crystal geometry in a DT ice layer. L is length of a single crystal facet. t is crystal thickness.

CONCLUSIONS

We have presented the first results from beta layering in 1 mm plastic shells with fill tubes. Our best beta layers have a roughness as small as 1.2 microns rms for a 210 μm thick ice layer. Layer smoothness depends upon cooling rate as well as layer thickness. Our best D-T ice layers are formed when freezing through the triple point is slower than 3 mK/min. for 210 μm layers. Data and simple calculations indicate that thinner layers are rougher layers.

ACKNOWLEDGMENTS

REFERENCES

[1] John Lindl, "Development of the indirect-drive approach to inertial confinement fusion and the target physics basis for ignition and gain," *Phys. Plasmas* **2** (11), 3393 (1995)

[2] K. Kim, L. C. Mok, M. J. Erlenborn, and T. P. Bernat, "Non contact thermal gradient method for fabrication of uniform cryogenic inertial fusion target," *J. Vac. Sci. Technol.* A. 1(2), p. 1196. (1985)

[3] J. R. Miller, Los Alamos Scientific Laboratory Report LA-6245-PR, Dec. 1975, p. 82; *Methods and Apparatus for Producing Cryogenic Inertially Driven Fusion Targets*, U. S. Patent 4,292,340 (Jan. 1987)

[4] A. J. Martin, R. J. Simms, and D. L. Musinski, KMS Fusion, Inc. Report No. 1348 (1985, unpublished); A. J. Martin, R. J. Simms, and R. B. Jacobs, *J. Vac. Sci. Technol.* A **6** (3), 1885. (1988)

[5] J. K. Hoffer and L. R. Foreman, *Phys. Rev. Lett.* **60**, 1310 (1988).

[6] S. Hann, S. Pollaine, J. Lindl, et al., *ICF Quarterly Report* 3(2), Lawrence Livermore National Laboratory, Livermore, CA, UCRL-LR-105821-95-4, p. 81, (1995).

[7] E. R. Mapoles, J. Sater, J. Pipes, E. Monsler, "Smoothing of deuterium-tritium ice by electrical heating of the saturated vapor", Phys. Rev. E. **55**, 3473 (1997)

[8] P. Clark Souers, *Hydrogen Properties for Fusion Energy*, (University of California Press, Berkeley, Ca. 1986), p. 61.

[9] B. J. Kozioziemski, G. W. Collins, T. P. Bernat, "Crystal Growth and Roughening of Solid D_2", *Fusion Technology* **31**, 482 (1997).

[10] W. W. Mullins, "Theory of Thermal Grooving", *Journal of Applied Physics*. **28**, 333 (1957)